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# Two-Detector Mode MGA Analysis of Plutonium using a Single Ge Detector*)

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**Abstract**

Plutonium gamma-ray data analysis using MGA in the two-detector mode can provide information more refined than the gamma-ray analysis using MGA in the one-detector mode. Prior to the introduction of the new type of ORTEC coaxial detector, which has good resolution at 100-keV region and good efficiency at 1 MeV, the two-detector mode of the MGA could be used only with two separate HPGe detectors with appropriate characteristics. A recent study by us using small plutonium standards (less than 0.5g) suggests that this new detector indeed performed as well as combination of a high-resolution planar ("LEPS") detector and a coaxial detector together. In this study, the CBNM plutonium gamma-ray standards were used to test this detector's ability with MGA, when measuring several grams of plutonium.

**Introduction**

The gamma-ray multi-group analysis code MGA[1,2,3] developed at the Lawrence Livermore National Laboratory is widely used for non-destructive plutonium gamma-ray assay for isotopic information. This plutonium isotopic analysis code is unique in that it de-convolutes the complicated, 100-keV x ray and gamma-ray region to obtain the ratios of the Pu isotopes as shown in Figure 1. As a result, MGA can determine the relative abundance of the plutonium isotopes with accuracy better than 1% using a high-resolution planar germanium detector in a few minutes of counting time.

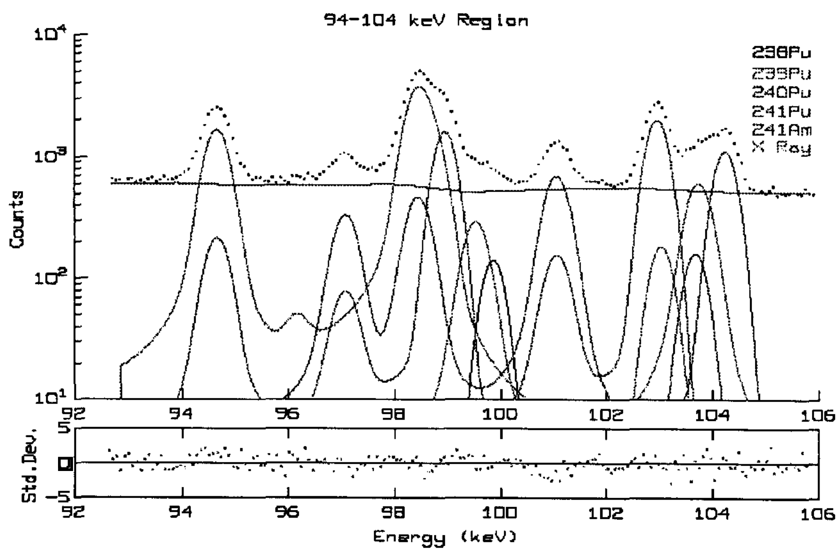

Figure 1. MGA analyzes the complicated 100-keV energy region to obtain plutonium-$^{241}$Am isotopic information

The code was developed to analyze either a low-energy gamma-ray spectrum taken using a high-resolution HPGe detector for energies below 300 keV, or to analyze a low-energy spectrum combined with a high-energy spectrum (up to 1 MeV) in what we refer to as the two-detector analysis mode. This was done originally to improve the determination of the $^{241}$Pu/$^{239}$Pu ratio in high burnup plutonium with the 300-keV regions from the high-energy spectrum. In high burnup plutonium, $^{241}$Pu is dominant in the 100-keV region and the precision of the $^{241}$Pu/$^{239}$Pu ratio from this region is reduced.

The need for two detectors to measure high burnup plutonium is inconvenient for inspection organizations, so MGA was upgraded to allow measurements of high burnup plutonium with a single high-resolution germanium detector. A rigorous determination of the relative detection efficiency for each measurement was developed that allowed the $^{241}$Pu/$^{239}$Pu ratio to be determined accurately from the 129- and 148-kev peaks of $^{239}$Pu and $^{241}$Pu, respectively. Eleven peaks in the low-energy region are used for this intrinsic efficiency calibration. Figure 2 illustrates how the relative detection efficiency for the measurement is determined for the low-energy detector.

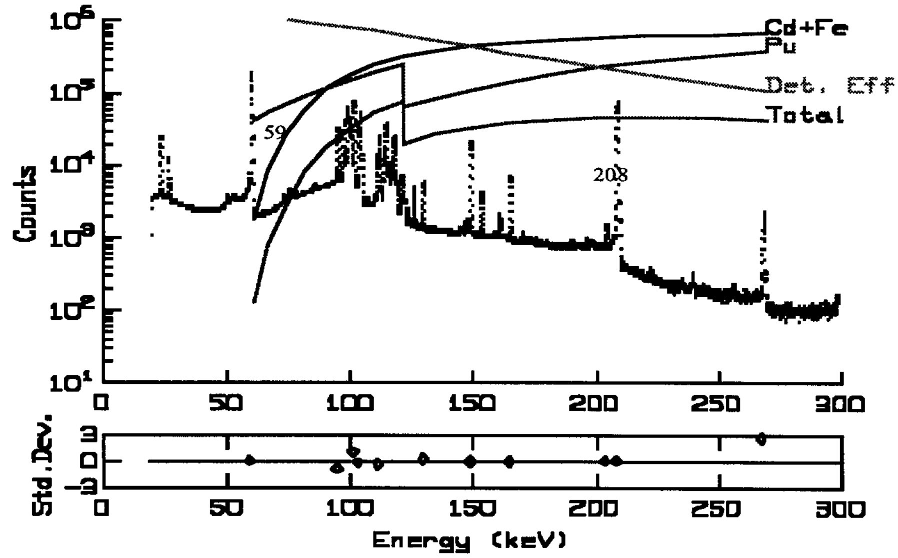


Figure 2. MGA uses physical attenuation corrections of both emission and absorption in the gamma ray interactions. This plot shows the three principal processes that characterize the low-energy "intrinsic" efficiency curve. The 59.5-, 94.6-, 101.1-, 103.0-, 11.0-, 129.3-, 148.6-, 164.6-, 203.5-, 208.0-, and 257.5-keV peaks are used in this characterization.

In general, MGA is now able to obtain accurate plutonium isotopic information for all burnups using the data collected with a high-resolution planar detector (below 300 keV). However, more refined isotopic results and additional isotopic information can be obtained from using higher-energy gamma rays (above 300 keV) collected with a high efficiency coaxial detector. For example, the $^{238}U$ abundance can be obtained from the 1001-keV peak, a more accurate analysis for $^{237}Np$ can be made using the 312-keV peak from $^{233}Pa$, some fission products such as $^{137}Cs$ and $^{95}Zr$-$^{95}Nb$ can be identified, and $^{241}Am$ inhomogeneities in the sample can also be determined by comparing the $^{241}Am/^{239}Pu$ ratio from different regions 9100-, 300-, and 600-keV) of the spectra. The two-detector mode of MGA is the only way to provide homogeneity information for a sample.

It is worth noting that these two sets of data do not have to be collected at the same time under the same geometry, because in each case an intrinsic efficiency curve is derived from the spectrum itself. To properly use the higher energy gamma-ray information, a separate intrinsic efficiency curve must be determined, as shown in Fig. 3. Like the low-energy curve in Fig. 2, the components of the efficiency are based on the physical processes involved in attenuating and detecting the gamma rays. In general, there is a thin Pb absorber in front of the coaxial detector to reduce the count rate due to the strong 100-keV region gamma rays.

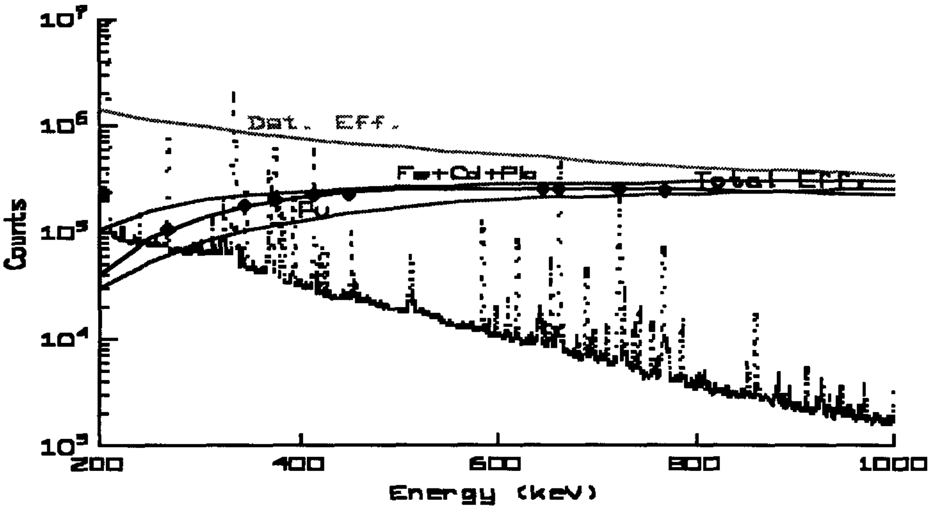


Figure 3. The same physical attenuation processes are used in analyzing the second detector data when running MGA in the two-detector mode. This plot shows the components affecting the efficiency curve. The Fe and Cd shown in the plot represent the steel wall of the container and the cadmium typically used behind the lead absorber to reduce the Pb x-rays.

The use of two detectors has been mandated by their conflicting requirements: a detector with excellent resolution at low energies (characteristic of small planar detectors) and a detector with good high-energy efficiency (characteristic of coaxial detectors). Usually, a high-energy spectrum taken using a coaxial HPGe detector will not provide sufficient energy resolution for 100-keV plutonium isotopic analysis, while the small planar HPGe used at low energies has inadequate high-energy efficiency. Prior to the introduction of the new ORTEC HPGe detector, the two-detector mode of the MGA could be used only with two separate Ge detectors having appropriate characteristics. They could be two separate Ge detectors or two Ge crystals mounted in “telescope” form within a single detector endcap. Telescope configurations suffer from the following disadvantages: the detector is actually "two in one." Failure of either detector means failure of both. Telescopes are complex to manufacture. Any filter inserted between the two crystals is internal to the detector and cannot practically be changed for different applications.

An optimized-geometry ORTEC HPGe detector has been developed which combines good energy resolution at 100 keV as well as acceptable high-energy (~1MeV) efficiency in a single detector. It can be used to gather spectra of both low- and high-energy regions of plutonium spectra simultaneously, for analysis by MGA in the two-detector mode. The detector is nominally 50mm diameter by 30 mm deep. The resolution at 122 keV is about 725 eV for a 2 μs shaping time at a count rate of 30 kHz. Applying the usual coaxial efficiency definition, the relative efficiency at 1.33MeV is approximately 15%, which is not too far away from the MGA suggested 20% coaxial detector efficiency.

In this paper we report on the MGA results with such a detector using CBNM plutonium standards (~6g in weight) with a $^{239}$Pu isotopic range from 60% to 90% for a detailed performance evaluation of this new detector. The data were collected using an ORTEC DSPEC spectrum analyzer at 0.075 keV/channel for 16k channels. The first 4k channel of the spectrum served as the "planar" data in the MGA two-detector mode analysis.

**The Detector**

Recently, three new ORTEC coaxial safeguard detectors were introduced; their specifications are listed in Table I. These detectors are optimized to have adequate resolution for low-energy gamma rays and good efficiency for high-energy gamma rays.

Because of both resolution and dead-time considerations in plutonium/MGA applications, we chose the SGD-GEM-15170 for our test. This detector has the best low energy resolution and possesses reasonable dead time for Pu measurements in a high radiation environment. The detector is nominally 50mm diameter by 30 mm deep. The resolution at 122 keV is about 725eV for a 2μs shaping time at a count rate of 30 kcps. Applying the usual coaxial efficiency definition, the relative efficiency at 1.33MeV is approximately 15% which is not too far away from the MGA suggested 20% coaxial detector efficiency. Figure 4 shows a plutonium gamma-ray spectrum taken with this 15% detector compared to a regular planar LEPS detector.

Table I. Specifications of the three new ORTEC SGD Safeguards Coaxial Detectors

| SGD GEM Coaxial Safeguards Detectors | | | | | | |
|---|---|---|---|---|---|---|
| Model number | Nominal | Nominal | Energy | Warranted Resolution | | |
| | Active Diameter | Thickness (mm) | | @1kcps (6μS) | @1kcps (2μS) | @30kcps (2μS) |
| | (mm) | | | | | |
| SGD-GEM-15170 | 50 | 30 | 122 keV | 625eV | 675eV | 725eV |
| | | | 1.33 MeV | 1.70keV | 1.85keV | 2.05keV |
| SGD-GEM-25175 | 50 | 50 | 122keV | 750eV | 870eV | 880eV |
| | | | 1.33 MeV | 1.75keV | 1.95keV | 2.0keV |

| | | | | | | |
|---|---|---|---|---|---|---|
| SGD-GEM-50180 | 65 | 65 | 122keV | 800eV | 925eV | 950eV |
| | | | 1.33 MeV | 1.80keV | 2.05keV | 2.15keV |
| First two digits in model number (e.g., the "25" in SGD-GEM25175) define the nominal relative efficiency, according to the usual IEE standard at 1.33 MeV | | | | | | |
| Peak shape at all count rates <= 30KHZ and time constants from 2-6μS, FW0.2M/FWHM <= 2.75 made with ORTEC 672 or DSPEC/DSPEC Plus | | | | | | |

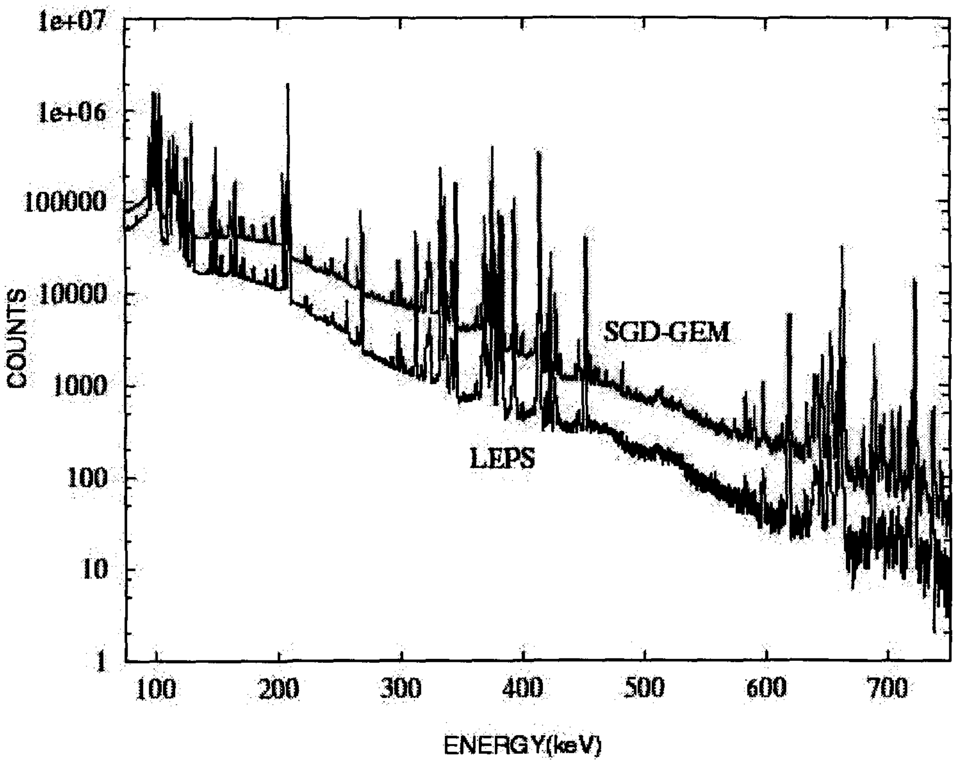


Figure 4. A comparison of plutonium gamma-ray spectra collected using the SGD-GEM detector (TOP) and a planar LEPS detector (Bottom). At 100-keV region, the LEPS has slightly better energy resolution, however, for higher energy (>100 keV) gamma rays, the SGD-GEM has significant higher efficiency. The source was placed at the same distance from both detectors and collected with the same live time.

**Experimental Setup**

The four CBNM standards were used in this study. These well-characterized sources have $^{240}Pu$ enrichment ranging from 6% to 25% and contain about 6.6 grams of $PuO_2$. Cadmium absorbers ranging from 30 mils to 50 mils in thickness were put between the source and the detector to reduce the count rate due to low energy Pu and 59-keV $^{241}Am$ gamma rays. Signals were collected using an ORTEC DSPEC digital spectrometer at 0.075 keV/channel for 16k channels. Data were collected for 1 hour, 6 hours, and 12 hours from the SGD-GEM detector.

**MGA analysis and results**

In the single detector mode, MGA uses the 100-keV region to obtain the Pu isotopic information as well as the $^{241}Am/Pu$ information (see Fig 1). The 100-keV region also provides the U/Pu ratio using fluorescence x-rays, the $^{235}U/Pu$ ratio is obtained by using the intensity of the 185-keV gamma rays. In the two-detector mode, MGA goes further in its analyses. Additional $^{241}Am/^{239}Pu$ ratios can be obtained by analyzing the gamma rays at 300-keV and 600-keV regions and these ratios along with the $^{241}Am/^{239}Pu$ ratio from the 100-keV region are compared to look for inhomogeneity in $^{241}Am$. MGA will indicate that inhomogeneity in $^{241}Am$ has been detected, however, the user needs to interpret these results. The $^{238}Pu/^{238}U$ ratio can be obtained using the 766 keV and the 1001 keV gamma rays. The $^{237}Np$ can be obtained using the 312 keV peak from the decay of the $^{233}Pa$ (the daughter product of $^{237}Np$.) A further check on the presence of uranium is performed using both the information of the uranium fluorescence x-rays from the analysis of the 100-keV region with the uranium information from the second detector. It is also worth noting that the MGA analysis in the two-detector mode can analyze MOX samples with U/Pu ratios ranging from 2 to 150.

The first 4k channels of the SGD-GEM data were used as the planar detector data for the one-detector mode MGA analysis or as the "first detector" (i.e., the "LEPS") data in the two-detector mode MGA analysis. The same data (up to 16k channels) were binned to 0.3keV/channel as the "second detector" (i.e., the "COAX" detector) data for MGA analysis in the two-detector mode. Table 3 shows all four of the CBNM sources results using the SGD-GEM detector in one detector mode, two-detector mode, compare to the mass-spectrometry numbers and a standard LEPS, the resolution at 100 keV for SGD-GEM is about 700 eV and 550 eV for LEPS.

Table 3. MGA analysis results from all four CBNM standards, when using the SGD-GEM data in the one-detector and two-detector modes. The number in brackets in each case is the % uncertainty on the value. The isotopic composition of the plutonium in CBNM is listed along with the $^{241}Am/Pu$ and $^{237}Np/Pu$ ratios. Low level $^{237}Np$ has been detected only in the two-detector mode.

| CBNM93 | SGD-GEM | SGD-GEM | LEPS | Declared Value |
|---|---|---|---|---|
| | One-detector | Two-detector | | |
| | | | | |
| Pu238 | 0.001280(1.93) | 0.001274(2.03) | 0.0001306(3.91) | 0.000117(.25) |
| Pu239 | 0.932504(.02) | 0.932593(.02) | 0.932163(.05) | 0.934123(0.0044) |
| Pu240 | 0.0647696(.32) | 0.0646868(.34) | 0.0650644(.68) | 0.063131(.062) |
| Pu241 | 0.0022034(.18) | 0.002198(.19) | 0.0022443(.40) | 0.002235(.13) |
| Pu242 | 0.00395(1.0) | 0.00395(1.0) | 0.00395(1.0) | 0.000395(2.5) |
| Am241 | 0.0010614(.53) | 0.001058(.39) | 0.0010406(.73) | 0.001039(2.2) |
| Np237 | | 0.00019(8.68) | | |

| CBNM84 | SGD-GEM | SGD-GEM | LEPS | Declared Value |
|---|---|---|---|---|
| | One-detector | Two-detector | | |
| | | | | |

| | | | | |
|---|---|---|---|---|
| Pu238 | 0.000709(1.25) | 0.0007066(1.29) | 0.0006983(1.1) | 0.000703(.85) |
| Pu239 | 0.840075(.10) | 0.841709(.09) | 0.842717(.20) | 0.843377(.001) |
| Pu240 | 0.145312(.34) | 0.1448377(.50) | 0.1429214(.70) | 0.142069(.056) |
| Pu241 | 0.0103258(.31) | 0.0103087(.30) | 0.0100873(.55) | 0.010275(.175) |
| Pu242 | 0.003576(1.0) | 0.003576(1.0) | 0.003576(1.0) | 0.003576(.28) |
| Am241 | 0.023071(1.20) | 0.0228(1.23) | 0.00242(1.32) | 0.02173(1.0) |
| Np237 | | 0.000106(2.47) | | |

| CBNM70 | SGD-GEM | SGD-GEM | LEPS | Declared Value |
|---|---|---|---|---|
| | One-detector | Two-detector | | |
| | | | | |
| Pu238 | 0.0086005(.69) | 0.085108(.50) | 0.085751(.81) | 0.008458(.21) |
| Pu239 | 0.730212(.28) | 0.73149(.21) | 0.7334839(.30) | 0.733191(.013) |
| Pu240 | 0.1861569(.34) | 0.185105(.81) | 0.1841726(1.02) | 0.182945(.048) |
| Pu241 | 0.0542586(.31) | 0.0536701(.34) | 0.053695(.70) | 0.054634(.062) |
| Pu242 | 0.020772(1.0) | 0.020772(1.0) | 0.020772(1.0) | 0.020772(.11) |
| Am241 | 0.011975(2.47) | 0.0118566(1.36) | 0.012654(2.63) | 0.011705(1.0) |
| Np237 | | 0.000687(2.07) | | |
| CBNM61 | SGD-GEM | SGD-GEM | LEPS | Declared Value |
| | One-detector | Two-detector | | |
| | | | | |
| Pu238 | 0.012101(.81) | 0.0121368(.51) | 0.0012052(.82) | 0.01969(.2) |
| Pu239 | 0.626072(.48) | 0.6249114(.29) | 0.6229547(.48) | 0.625255(.045) |
| Pu240 | 0.256394(1.05) | 0.2573618(.77) | 0.2577372(.93) | 0.254058(.095) |
| Pu241 | 0.063501(.73) | 0.0636651(.36) | 0.0653269(.75) | 0.066793(.13) |
| Pu242 | 0.041925(1.0) | 0.041925(1.0) | 0.041925(1.0) | 0.41925(.15) |
| Am241 | 0.016785(2.5) | 0.016102(2.6) | 0.0160479(2.71) | 0.014452(0.9) |
| Np237 | | 0.00863(1.89) | | |

**Conclusions**

Because of its adequate energy resolution at 100-keV region and good efficiency at 1 MeV (~15%), the SGD-GEM detector performs as well as a LEPS detector when using MGA in the one-detector mode for data analysis. The gamma-ray spectrum collected with the SGD-GEM detector contains far better isotopic information (e.g., $^{237}$Np, $^{238}$U/$^{235}$U, etc.) than the spectrum collected with a LEPS detector when using MGA in the two-detector mode for data analysis. We have shown that MGA can analyze the data collected with such a detector and a DSPEC type analyzer in either one detector or two-detector mode. Thus ORTEC SGD-GEM type detector can be a powerful tool when combining the two-detector mode MGA for analyzing U, Pu mixtures, MOX samples, and Pu[4)] immobilized in ceramic. The detector can also be used with MGA4U[5], MGAHI[6] isotopic analysis software for uranium, shielded-plutonium data analysis, respectively.

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